

Charge conservation breaking within generalized master equation description of electronic transport through dissipative double quantum dots

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Abstract

We report an observation of charge conservation breaking in a model study of electronic current noise of transport through a dissipative double quantum dot within generalized master equation formalism. We study the current noise through a double quantum dot coupled to two electronic leads in the high bias limit and a dissipative heat bath in the weak coupling limit. Our calculations are based on the solution of a Markovian generalized master equation. Zero-frequency component of the current noise calculated within the system, i.e., between the two dots, via the quantum regression theorem exhibits unphysical negative values. On the other hand, current noise calculated for currents between the dots and the leads by the counting variable approach shows no anomalies and seems physically plausible. We inquire into the origin of this discrepancy between two nominally equivalent approaches and show that it stems from the simultaneous presence of the two types of baths, i.e., the electronic leads and the dissipative bosonic bath. This finding raises interesting questions concerning conceptual foundations of the theory describing multiple-baths open quantum systems widely encountered in nanoscience.

Key words: generalized master equation, quantum Markov processes, charge conservation, dissipation
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1. Introduction

Recent advances in technology, fabrication, and measurement of mesoscopic semiconductor devices with ever-decreasing dimensions of achievable nanostructures stimulate also theoretical studies of physical phenomena determining their properties. Questions of prime interest concern their possible quantum behavior and the quantum-classical crossover due to interaction with surrounding environment causing dissipation, relaxation, and dephasing [1–3]. One of the conceptually simplest and experimentally achieved

systems is the double quantum dot (DQD) which can be tuned into a regime where it is effectively described as a tunable two-level system for the electronic energy states [4]. These energy states can be tuned, for instance, by means of an external gate voltage. The interest in such devices stems also from the attractive possibility to utilize them as potential q-bits.

In the setup consisting of a DQD, the role of the electronic coherence between the two spatially separated electronic states corresponding to the respective dots is of central importance. The DQD device loses coherence due to the coupling to noisy environment (e.g. noise in the gate voltages and unavoidable interaction with phonons in the substrate). Moreover, energy can

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be exchanged with bosonic degrees of freedom which gives rise to transitions between states of nonequal energy and, thus, relaxation. The dissipative dynamics of two level systems (spin-boson problem) have been subject of study for many years [1,5]. DQD setup brings along a new twist to this standard problem in the fact that the DQD is electrically contacted by leads and charge transport through the DQD occurs [4]. This is a new feature of the old problem adding complexity to the methods of its solution. On the other hand, for most practically interesting setups the dissipative coupling may be considered as rather weak which should bring about important simplifications for its solution. Yet, as we will demonstrate in this work, the simultaneous presence of the two different kinds of baths (electronic leads and standard dissipative bosonic bath à la Caldeira-Leggett [6]) causes serious conceptual problems within the simplest possible formalism of Markovian generalized master equations (GME) when it is applied to the weakly dissipative DQD problem.

It has been suggested by Aguado and Brandes [7] that considering the electronic current noise in the DQD devices may be a useful tool for characterizing their dissipative properties going beyond the information available from the stationary current characteristics only. In this work we adopt their model and perform an exhaustive comparative study of the evaluation of the current noise based on GME approach used in previous studies [7,8]. We surprisingly find an internal inconsistency of the formalism which breaches fundamental physical law of charge conservation. Two nominally equivalent approaches for the calculation of the zero-frequency component of the current noise spectrum, namely the quantum regression theorem (QRT) and counting variable approach with the MacDonald formula, show mutual discrepancy. This fact puts under question the status of the results obtained by these methods.

The structure of the paper is the following. In Sec. 2 we introduce our model and in Sec. 3 we describe the method of its solution via an approximate Markovian GME. The following Sec. 4 describes the evaluation of the current noise within the used GME formalism while Sec. 5 discusses the general aspects of the charge conservation and its breaking within the GME formalism. Sec. 6 presents an overview of results for our studied model of a dissipative DQD. We discuss the obtained results and their implications together with resulting

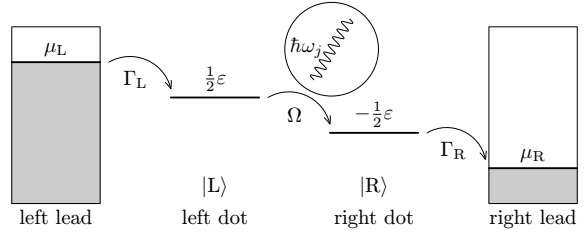


Fig. 1. Schematic depiction of the studied DQD system.

open problems and outlook in the concluding Sec. 7.

2. Model

The double quantum dot device [4] in Fig. 1 is described as two electronic levels (corresponding to particular single-electron levels within the transport window of the left and right dot, respectively) de-aligned by an energy difference ε with a coherent interdot tunnel coupling Ω . The system is assumed to be in the regime of strong Coulomb blockade so that only three states play a role: no extra electron $|0\rangle$ on the whole DQD system, one extra electron on the left dot $|L\rangle$ and one extra electron on the right dot $|R\rangle$, i.e., we exclude multiple occupancies of the DQD. This can be achieved by a suitable gating, when a very high charging energy prohibits an addition of more than one electron. We also consider spinless electrons. Hamiltonian of the DQD device then reads

$$H_S = \frac{1}{2}\varepsilon(|L\rangle\langle L| - |R\rangle\langle R|) + \Omega(|L\rangle\langle R| + |R\rangle\langle L|). \quad (1)$$

The so called device bias ε can be tuned by gating. The term proportional to Ω enables the tunnel current through the device. The eigenvalues of the isolated system Hamiltonian are $E_{1,2} = \pm\frac{1}{2}\Delta$ with $\Delta = \sqrt{4\Omega^2 + \varepsilon^2}$ and the corresponding eigenvectors read $|1\rangle = \sqrt{\frac{\Delta+\varepsilon}{2\Delta}}|L\rangle + \sqrt{\frac{\Delta-\varepsilon}{2\Delta}}|R\rangle$, $|2\rangle = -\sqrt{\frac{\Delta-\varepsilon}{2\Delta}}|L\rangle + \sqrt{\frac{\Delta+\varepsilon}{2\Delta}}|R\rangle$.

The double quantum dot is coupled to two leads with a high bias applied between them. The bias is smaller than the charging energy but otherwise it is the largest energy scale in the model. We assume that the non-interacting leads are coupled via standard tunneling terms

$$\begin{aligned}
H_C + H_{CS} = & \sum_k E_{kL} c_{kL}^\dagger c_{kL} + \sum_k E_{kR} c_{kR}^\dagger c_{kR} \\
& + \sum_k V_{kL} (c_{kL}^\dagger |0\rangle \langle L| + |L\rangle \langle 0| c_{kL}) \\
& + \sum_k V_{kR} (c_{kR}^\dagger |0\rangle \langle R| + |R\rangle \langle 0| c_{kR}).
\end{aligned} \quad (2)$$

The leads are held at respective electrochemical potentials μ_L and μ_R whose difference gives the bias. We assume that $\mu_L \rightarrow \infty$ and $\mu_R \rightarrow -\infty$. The tunneling densities of states $\Gamma_\alpha(\varepsilon) = \frac{2\pi}{\hbar} \sum_k |V_{k\alpha}|^2 \delta(\varepsilon - E_{k\alpha})$, $\alpha = L, R$ are assumed energy independent (wide-band limit or first Markov approximation, compare Ref. [2]) and equal $\Gamma_L = \Gamma_R = \Gamma$. Both the high bias and wide-band limits are necessary for the applicability of the Markov approximation later on.

Finally, we introduce a generic dissipative heat bath à la Caldeira-Leggett consisting of an infinite set of harmonic oscillators [1,6] which are linearly coupled to the left-right population difference of the DQD [7]

$$\begin{aligned}
H_B + H_{BS} = & \sum_j \hbar \omega_j (a_j^\dagger a_j + \frac{1}{2}) \\
& + \sum_j C_j (a_j^\dagger + a_j) (|L\rangle \langle L| - |R\rangle \langle R|).
\end{aligned} \quad (3)$$

The heat bath is fully characterized by its spectral density

$$J(\omega) = 2 \sum_j |C_j|^2 \delta(\omega - \omega_j), \quad (4)$$

which we take in the Ohmic form $J(\omega) = 2\hbar^2 \gamma \omega / \pi \cdot \exp(-\omega/\omega_c)$. The parameter γ gives the strength of the dissipation and ω_c is a high energy cut-off frequency [1,6].

3. Generalized master equation

3.1. Liouville space

In order to conveniently manipulate with density operators, we define Liouville space [8]. The Liouville space is a linear space spanned over operators acting on the original Hilbert space assigned to the system. Its basis $|n, n'\rangle\rangle$ is constructed from a basis $|n\rangle$ of the Hilbert space as $|n, n'\rangle\rangle \equiv |n\rangle \langle n'|$. A general operator in the Hilbert space

$$A = \sum_{n, n'} A_{nn'} |n\rangle \langle n'|$$

corresponds to the vector

$$|A\rangle\rangle = \sum_{n, n'} A_{nn'} |n, n'\rangle\rangle$$

in the Liouville space. For example, a density operator ρ can be written as $\rho = \sum_{n, n'} \rho_{nn'} |n, n'\rangle\rangle$ with $\rho_{nn'} = \langle n | \rho | n' \rangle$. We define the scalar product on the Liouville space by $\langle\langle A | B \rangle\rangle \equiv \text{Tr}\{A^\dagger B\}$. In order to avoid confusion, linear operators acting in the Liouville space are called superoperators and in the following will be denoted by calligraphic symbols. The vectors of the Liouville space in the bra-ket notation will be distinguished by double brackets. The matrix representation of superoperators then follows from the previous

$$\mathcal{A} = \sum_{\substack{n, n' \\ m, m'}} A_{nn', mm'} |n, n'\rangle\rangle \langle\langle m, m'|.$$

3.2. Liouvillean

For the description of a dissipative system we distinguish between the system itself (electronic states of the dots) and the reservoirs (heat bath and leads). Our task now is to get a closed evolution equation for the reduced density operator which is the system part only of the total density operator. To this end we perform the standard projection onto the system assuming weak coupling to the reservoirs and consequently using the Markov approximation [2]. Due to the weak coupling the effects of the two baths are additive.

It should be noted that within our assumptions on the leads (wide band limit and high bias) an equivalent result for the effect of leads can be obtained without the weak coupling assumption, i.e., for arbitrary Γ , as shown by Gurvitz and Prager [9]. It turns out that these assumptions correspond exactly to the so called singular coupling limit in the mathematical literature (see, e.g., Ref. [10]) which also leads to a Markovian dissipative evolution of the system. The additivity of the two bath is then, however, only a heuristic assumption which may break down for large enough Γ . The range of parameters we consider is restricted to rather small Γ so that the potential differences between the two possible approaches should be safely negligible and the result of the projection leads to the following GME

$$\frac{d\rho(t)}{dt} = \mathcal{L}\rho(t) = \mathcal{L}_S\rho(t) + \mathcal{L}_B\rho(t) + \mathcal{L}_C\rho(t), \quad (5)$$

with

$$\begin{aligned}
\mathcal{L}_S \rho(t) &= -\frac{i}{\hbar} [H_S, \rho(t)], \\
\mathcal{L}_B \rho(t) &= -\frac{1}{\hbar^2} \int_0^\infty d\tau \\
&\quad \times \text{Tr}_B \{ [H_{BS}, [H_{BS}(-\tau), \rho(t) \otimes \rho_B]] \}, \\
\mathcal{L}_C \rho(t) &= -\frac{1}{\hbar^2} \int_0^\infty d\tau \\
&\quad \times \text{Tr}_C \{ [H_{CS}, [H_{CS}(-\tau), \rho(t) \otimes \rho_C]] \}.
\end{aligned}$$

The part \mathcal{L}_S describes the free evolution of the system while $\mathcal{L}_B, \mathcal{L}_C$ determine the dissipative influence of the generic heat bath and the electronic leads, respectively. It turns out that the off-diagonal elements ρ_{0k}, ρ_{k0} with $k = 1, 2$ of the reduced density matrix are decoupled from the rest of the system, i.e., their evolution does not enter expressions for the other matrix elements and vice versa (compare with Ref. [8]). Therefore, the subspace $\{|0\alpha\rangle, |\alpha 0\rangle\}$ ($\alpha = L, R$) can be projected out leaving us with the relevant Liouville subspace with the basis $\{|00\rangle, |LL\rangle, |RR\rangle, |RL\rangle, |LR\rangle\}$. In this basis the above parts of the total Liouvillian are described by the following matrices [7,11]

$$\mathcal{L}_S = \frac{1}{\hbar} \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -i\Omega & i\Omega \\ 0 & 0 & 0 & i\Omega & -i\Omega \\ 0 & -i\Omega & i\Omega & i\epsilon & 0 \\ 0 & i\Omega & -i\Omega & 0 & -i\epsilon \end{pmatrix}, \quad (6)$$

$$\mathcal{L}_B = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & \gamma_+ & -\gamma_- & -\gamma_p & 0 \\ 0 & \gamma_+ & -\gamma_- & 0 & -\gamma_p \end{pmatrix}, \quad (7)$$

$$\mathcal{L}_C = \begin{pmatrix} -\Gamma & 0 & \Gamma & 0 & 0 \\ \Gamma & 0 & 0 & 0 & 0 \\ 0 & 0 & -\Gamma & 0 & 0 \\ 0 & 0 & 0 & -\frac{1}{2}\Gamma & 0 \\ 0 & 0 & 0 & 0 & -\frac{1}{2}\Gamma \end{pmatrix}, \quad (8)$$

with

$$\gamma_\pm = -\frac{\pi}{\hbar^2} \frac{\Omega}{\Delta} J(\Delta/\hbar) \left[\frac{\epsilon}{\Delta} \coth(\frac{1}{2}\beta\Delta) \pm 1 \right], \quad (9)$$

$$\gamma_p = \frac{4\pi}{\hbar^2} \frac{\Omega^2}{\Delta^2} J(\Delta/\hbar) \coth(\frac{1}{2}\beta\Delta). \quad (10)$$

4. Current noise

In this section we briefly introduce the current noise [12,13] and methods of its evaluation within the GME framework. More detailed account of these issues can be found in Sec. III of Ref. [8].

Equations of motion for the operators of the occupation of the left dot $n_L = |L\rangle\langle L|$ and the right dot $n_R = |R\rangle\langle R|$ read

$$e \frac{d}{dt} n_L = -\frac{ie}{\hbar} [n_L, H] = I_{L0} - I_{RL}, \quad (11)$$

$$e \frac{d}{dt} n_R = -\frac{ie}{\hbar} [n_R, H] = I_{RL} - I_{0R}. \quad (12)$$

On the right side of the equations, we identify charge current operators across the different junctions: $I_{L0} = -\frac{ie}{\hbar} [n_L, H_{CS}]$ is the operator of the current between the left lead and the left dot, $I_{RL} = \frac{ie}{\hbar} [n_L, H_S] = -\frac{ie}{\hbar} [n_R, H_S]$ is the operator of the current between the dots, and $I_{0R} = \frac{ie}{\hbar} [n_R, H_{CS}]$ is the operator of the current between the right dot and the right lead. Explicitly, they read

$$I_{L0} = \frac{ie}{\hbar} \sum_k V_{kL} (c_{kL}^\dagger |0\rangle\langle L| - |L\rangle\langle 0| c_{kL}), \quad (13)$$

$$I_{RL} = \frac{ie}{\hbar} \Omega (|L\rangle\langle R| - |R\rangle\langle L|), \quad (14)$$

$$I_{0R} = \frac{ie}{\hbar} \sum_k V_{kR} (|R\rangle\langle 0| c_{kR} - c_{kR}^\dagger |0\rangle\langle R|). \quad (15)$$

Since the commutators with the bath operators are zero $[n_L, H_{BS}] = [n_R, H_{BS}] = 0$, the heat bath gives no explicit contribution to the current operators.

The current operator I_{RL} is obviously a system operator, i.e., it acts as unity on the degrees of freedom of the leads and the heat bath. However, this is not the case for the two operators of current between the dots and the leads I_{L0} and I_{0R} .

Next, we define the current autocorrelation function

$$\begin{aligned}
C_A(\tau) \equiv \lim_{t \rightarrow \infty} \left[\frac{1}{2} \langle \{I_A(t+\tau), I_A(t)\} \rangle \right. \\
\left. - \langle I_A(t+\tau) \rangle \langle I_A(t) \rangle \right], \quad (16)
\end{aligned}$$

with $A = L0, RL, 0R$. Due to the stationary limit ($t \rightarrow \infty$) the autocorrelation function is symmetric $C_A(\tau) = C_A(-\tau)$. We define the current noise spectrum as

$$S_A(\omega) \equiv \int_{-\infty}^{\infty} d\tau C_A(\tau) e^{i\omega\tau}. \quad (17)$$

The current noise spectrum is non-negative as can be shown by using the Lehmann representation.

Now we need to express the current noise spectrum in terms of the quantities involved in the GME (5). We denote the stationary reduced density matrix $\lim_{t \rightarrow \infty} \rho(t) = \rho_{\text{stat}} \equiv |0\rangle$. It satisfies $\mathcal{L}\rho_{\text{stat}} = 0$, hence it is the zero-eigenvalue (right) eigenstate of the Liouvillian. Since the Liouvillian is not Hermitian, left zero-eigenvalue eigenstate denoted by $\langle\tilde{0}|$ is not just the Hermitian conjugate of the right zero-eigenvalue eigenstate $|0\rangle$. However, one can see that $\langle\tilde{0}| \equiv 1$, because for an arbitrary system operator A

$$\langle\tilde{0}|\mathcal{L}|A\rangle = \text{Tr}_S(1\mathcal{L}A) = \text{Tr}_S(\mathcal{L}A) = 0$$

due to normalization of the reduced density matrix. Now, we define the projector on the kernel of the Liouvillian $\mathcal{P} \equiv |0\rangle\langle\tilde{0}|$ and its orthogonal complement $\mathcal{Q} \equiv 1 - \mathcal{P}$. With help of \mathcal{Q} the well-defined superoperator $\mathcal{R} \equiv \mathcal{Q}\mathcal{L}^{-1}\mathcal{Q}$ represents the pseudoinverse of the Liouvillian (“inverse on the regular subspace \mathcal{Q} ”).

With that we have all necessary ingredients for expressing the current noise. It can be done in two different ways for the two types of junctions. The RL-junction lies in the system and, thus, the quantum regression theorem (see Ref. [2], Sec. 5.2) can be used to calculate the correlation function $C_{\text{RL}}(\tau)$. The final formula for the zero-frequency current noise reads [8]

$$S_{\text{RL}}(0) = -2e^2 \langle\tilde{0}|\mathcal{I}_{\text{RL}}\mathcal{R}\mathcal{I}_{\text{RL}}|0\rangle, \quad (18)$$

where we have introduced the current superoperator

$$\mathcal{I}_{\text{RL}}\rho \equiv \frac{1}{2e} \{I_{\text{RL}}, \rho\} \quad (19)$$

in terms of which the stationary current is given as $\langle I_{\text{RL}} \rangle = \text{Tr}_S(I_{\text{RL}}\rho_{\text{stat}}) = e\langle\tilde{0}|\mathcal{I}_{\text{RL}}|0\rangle$.

For the outer junctions (between the dots and the leads) the QRT cannot be used, because the current operators I_{L0} and I_{0R} involve the lead operators. However, n -resolved form of the generalized master equation and the MacDonald formula [13,14] enables us to calculate the zero-frequency noise also for these junctions, see details in Ref. [8], Sec. III. For the stationary mean current through the 0R-junction we get $\langle I_{\text{0R}} \rangle = e \text{Tr}_S(\mathcal{I}_{\text{0R}}\rho_{\text{stat}}) = e\langle\tilde{0}|\mathcal{I}_{\text{0R}}|0\rangle$ with

$$\mathcal{I}_{\text{0R}}\rho = \Gamma|0\rangle\langle\text{R}|\rho|\text{R}\rangle\langle 0|. \quad (20)$$

The final result for the zero-frequency current noise reads

$$S_{\text{0R}}(0) = e^2 \langle\tilde{0}|\mathcal{I}_{\text{0R}} - 2\mathcal{I}_{\text{0R}}\mathcal{R}\mathcal{I}_{\text{0R}}|0\rangle \quad (21)$$

and analogously for the L0-junction with $\mathcal{I}_{\text{L0}}\rho = \Gamma|\text{L}\rangle\langle 0|\rho|0\rangle\langle\text{L}|$.

5. Charge conservation issue

The equations of motion (11) and (12) for the dot occupation operators (charge conservation conditions) imply that the stationary mean current and the zero frequency noise are independent of the measurement position along the circuit [8]

$$\langle I_{\text{L0}} \rangle = \langle I_{\text{RL}} \rangle = \langle I_{\text{0R}} \rangle, \quad S_{\text{L0}}(0) = S_{\text{RL}}(0) = S_{\text{0R}}(0).$$

Let us now focus on the reformulation of the charge conservation condition in the superoperator language and evaluate, e.g., the commutator $[\mathcal{N}_{\text{L}}, \mathcal{L}]$ with the superoperator of occupation of the left dot defined by $\mathcal{N}_{\text{L}}\rho = \frac{1}{2}\{n_{\text{L}}, \rho\}$ in analogy with other superoperators corresponding to system operators such as, e.g., \mathcal{I}_{RL} . Its matrix representation in the relevant 5-dimensional Liouville subspace reads $\mathcal{N}_{\text{L}} = \text{diag}(0, 1, 0, 1/2, 1/2)$. Then with the help of Eqs. (6), (7), and (8) we arrive at

$$[\mathcal{N}_{\text{L}}, \mathcal{L}] = -\mathcal{I}_{\text{RL}} - \mathcal{I}_{\text{A}} + \mathcal{I}_{\text{L0}}, \quad (22)$$

with the “anomalous current superoperator” $\mathcal{I}_{\text{A}} = -[\mathcal{N}_{\text{L}}, \mathcal{L}_{\text{B}}]$. Since $[n_{\text{L}}, H_{\text{BS}}] = 0$, we would expect that also $[\mathcal{N}_{\text{L}}, \mathcal{L}_{\text{B}}]$ is zero, therefore the final Liouville space analogy to the charge conservation condition (11) should read $[\mathcal{N}_{\text{L}}, \mathcal{L}] = \mathcal{I}_{\text{L0}} - \mathcal{I}_{\text{RL}}$. Indeed, if this condition were satisfied one could easily explicitly show the equivalence of Eqs. (18) and (21) using the method of Ref. [8], Sec. IIID. Unfortunately, as one can readily evaluate

$$\mathcal{I}_{\text{A}} = \frac{1}{2} \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & \gamma_+ & \gamma_- & 0 & 0 \\ 0 & \gamma_+ & \gamma_- & 0 & 0 \end{pmatrix}, \quad (23)$$

so that the anomalous current superoperator is not identically zero. Analogously, for the other dot we obtain the result

$$[\mathcal{N}_{\text{R}}, \mathcal{L}] = \mathcal{I}_{\text{RL}} + \mathcal{I}_{\text{A}} - \mathcal{I}_{\text{0R}}, \quad (24)$$

with the opposite contribution from the heat bath compared to the left dot, i.e., $[\mathcal{N}_{\text{R}}, \mathcal{L}_{\text{B}}] = -[\mathcal{N}_{\text{L}}, \mathcal{L}_{\text{B}}] = \mathcal{I}_{\text{A}}$.

Thus, we face the fact that the charge conservation condition contains nonzero anomalous terms. The possible consequences may be that the mean current or the zero-frequency current noise are no longer equal for all pairs of junctions. First, since $\langle\tilde{0}|$ corresponds to the unity operator and, thus, is equal to $(1, 1, 1, 0, 0)$,

we see that $\langle\langle\tilde{0}|\mathcal{I}_A|0\rangle\rangle = 0$ and the mean current is conserved along the whole circuit. Also, we notice that $[\mathcal{N}_L + \mathcal{N}_R, \mathcal{L}_B] = 0$ implies there is no problem with the charge conservation between the outer junctions. However, the zero-frequency noise eventually does show discrepancy between the outer junctions and the inner one, in particular

$$S_{RL}(0) - S_{L0}(0) = e^2 \langle\langle\tilde{0}|\mathcal{I}_{L0}\mathcal{R}\mathcal{I}_A|0\rangle\rangle = e^2 \Gamma \Omega \varepsilon \times \frac{\hbar^2 \Gamma \gamma_+ \Lambda^2 + 2\Omega^2 \Lambda (\gamma_+ + \gamma_-) - 2\varepsilon^2 \gamma_+ (\frac{2\Omega}{\varepsilon} \gamma_- - \frac{1}{2}\Gamma)}{[\frac{1}{2}\Gamma \varepsilon^2 + 3\Omega^2 \Lambda + \frac{1}{2}\hbar^2 \Gamma \Lambda^2 - \Omega \varepsilon (2\gamma_+ + \gamma_-)]^2}, \quad (25)$$

where $\Lambda = \gamma_p + \frac{1}{2}\Gamma$. We will analyze features of this discrepancy in detail in the next section.

Before that, let us return to the point where we have assumed that $[\mathcal{N}_L, \mathcal{L}_B]$ was identically zero. This was a reasonable assumption, because the dot occupation operator n_L commutes with the heat bath-system interaction Hamiltonian H_{BS} and thus the bath variables do not enter explicitly the current operators. It can be shown that $[\mathcal{N}_L, \mathcal{L}]$ indeed does not depend on the bath variables if we operate on the whole Liouville space (system + bath) before projecting onto the system and introducing the weak coupling and Markovian limits [15]. However, after the projection we arrive at

$$\begin{aligned} [\mathcal{N}_L, \mathcal{L}_B]\rho &= \\ &= \frac{1}{2\hbar^2} \int_0^\infty d\tau \text{Tr}_B \{ [H_{BS}, n_L], [H_{BS}(-\tau), \rho \otimes \rho_B] \} \\ &+ \frac{1}{2\hbar^2} \int_0^\infty d\tau \text{Tr}_B \{ [H_{BS}, \{ [H_{BS}(-\tau), n_L], \rho \otimes \rho_B \}] \}. \end{aligned} \quad (26)$$

Apparently, the first term is equal to zero due to $[n_L, H_{BS}] = 0$ as expected, while the second term yields the nonzero anomalous current.

6. Results

In this section we present various aspects of the previously found charge conservation breaking in the dissipative DQD model. In the following we will conventionally represent the noise by a dimensionless quantity, the Fano factor $F = S(0)/e\langle I \rangle$ [12].

6.1. Zero dissipation

In the case without the heat bath ($\gamma = 0$), the charge conservation condition is satisfied. For the mean current [16] and the Fano factor [14] we obtain

$$\langle I \rangle = e\Gamma \frac{\Omega^2}{\varepsilon^2 + 3\Omega^2 + (\frac{1}{2}\hbar\Gamma)^2},$$

$$F_{L0} = F_{RL} = F_{0R} = 1 - \frac{4\Omega^2 (\Omega^2 + 2(\frac{1}{2}\hbar\Gamma)^2)}{(\varepsilon^2 + 3\Omega^2 + (\frac{1}{2}\hbar\Gamma)^2)^2}.$$

These results are illustrated in Fig. 2. The mean current vs. bias ε has the Lorentzian shape with the half-width $\sqrt{3\Omega^2 + (\hbar\Gamma/2)^2}$ and maximum at $\varepsilon = 0$. The Fano factor has the dip at $\varepsilon = 0$ where quantum coherence strongly suppresses the noise. For large $|\varepsilon| > 0$ the mean current becomes very small and thus electrons tunnel very sparsely and consequently the tunneling events are uncorrelated which corresponds to a Poisson process with the value of the Fano factor $F \rightarrow 1$.

6.2. General case

When dissipative heat bath comes into play ($\gamma > 0$), the transport is strongly affected by the possibility of exchanging energy with the heat bath [7] as it is illustrated in Fig. 2. The shape of the mean current curve is no longer Lorentzian but exhibits an asymmetry. With increasing temperature the peak becomes broader and more symmetric. Analytically, we obtain

$$\langle I \rangle = \frac{e\Gamma\Omega (\Omega\Lambda - \gamma_+\varepsilon)}{\frac{1}{2}\Gamma\varepsilon^2 + 3\Omega^2\Lambda + \frac{1}{2}\hbar^2\Gamma\Lambda^2 - \Omega\varepsilon(2\gamma_+ + \gamma_-)}. \quad (27)$$

In the right plot of Fig. 2 Fano factors $F_{L0} = F_{0R}$ (solid lines) and F_{RL} (dotted lines) are plotted. The difference between the Fano factors obtained by different approaches is significant. Interestingly, at $\varepsilon = 0$ the Fano factors have the same value $F_{L0} = F_{RL}$ as follows from relation (25).

For $\varepsilon > 0$ spontaneous emission occurs even at very low temperatures and the noise is reduced well below the Poisson limit. Larger couplings γ lead to very asymmetric Fano factor. At finite temperatures, absorption of energy quanta from the bath is also possible and the Fano factor for $\varepsilon < 0$ is reduced below the Poisson limit too. With increasing temperature the effect of the emission and the absorption is growing, except the point $\varepsilon = 0$ where both the mean current

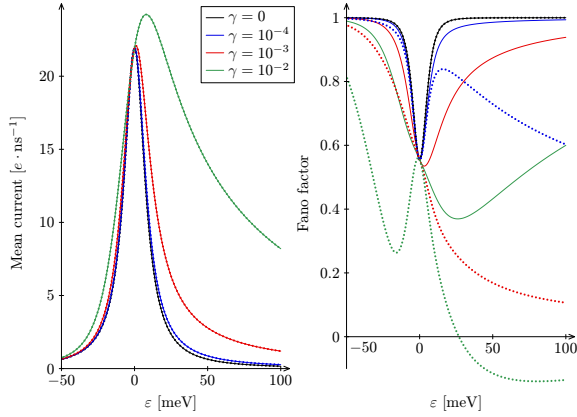


Fig. 2. Mean current and Fano factor vs. device bias ε for different values of damping coefficient γ . Solid lines show the mean current and the Fano factor at the outer junctions L0, 0R, dotted lines at the inner junction RL. Parameters: $\Omega = 5$ meV, $\Gamma = 0.1/\hbar$ meV, $\beta = 0.1$ meV $^{-1}$.

and the Fano factor are temperature independent. It appears that the MacDonald formula yields physically plausible results for F_{L0} and F_{0R} , whereas F_{RL} given by the quantum regression theorem behaves pathologically with unphysical negative values and non-Poisson limit for $\varepsilon \rightarrow \infty$. For sufficiently strong coupling ($\gamma \approx 10^{-2}$) F_{RL} drops to negative values in the $\varepsilon > 0$ region and for sufficiently high temperature ($T \approx 200$ K) also in the $\varepsilon < 0$ region. Analyzing the expression (25) we find that for $\varepsilon \rightarrow \infty$ the noise difference $\Delta S = S_{RL}(0) - S_{L0}(0) \sim 1/\varepsilon$. From the relation (27) for the mean current in the same limit it follows $\langle I \rangle \sim 1/\varepsilon$. Therefore their ratio yielding the difference of Fano factors $\Delta F = F_{RL} - F_{L0}$ does not go to zero for $\varepsilon \rightarrow \infty$ as it should. Nevertheless, despite of the fact that F_{RL} behaves manifestly wrong, we do not have any valid proof yet, that the MacDonald formula gives a better and more reliable results for F_{L0} and F_{0R} .

In the following subsections we will investigate how both the MacDonald formula and the quantum regression theorem approach behave in several approximations or limit cases. It will answer whether the MacDonald formula gives physically acceptable results and will show more pathologies of the quantum regression theorem results.

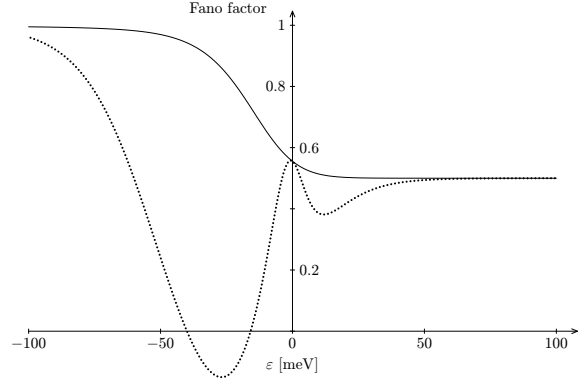


Fig. 3. Fano factor vs. device bias ε for $\Gamma \rightarrow 0$. Solid line shows $F_{L0} = F_{0R}$, dotted line F_{RL} . Parameters: $\Omega = 5$ meV, $\beta = 0.1$ meV $^{-1}$.

6.3. Limit $\Gamma \rightarrow 0$

Limit $\Gamma \rightarrow 0$ could be potentially interesting – analogously with the dissipationless limit $\gamma \rightarrow 0$ the charge conservation could possibly be recovered. The mean current and the zero-frequency noise go to zero in this limit, however their ratio, the Fano factor, does not [15]. The difference between the Fano factors of the outer and inner junctions reads

$$F_{L0} - F_{RL} = \frac{2\varepsilon^2 (\coth^2(\frac{1}{2}\beta\Delta) - 1)}{\varepsilon^2 + 4\varepsilon\Delta \coth(\frac{1}{2}\beta\Delta) + 3\Delta^2 \coth^2(\frac{1}{2}\beta\Delta)}.$$

Results are illustrated in Fig. 3. We note quite an interesting phenomenon that the Fano factor does not depend on the heat bath spectral density and, thus, it is not influenced by the strength γ of the dissipation. Nevertheless, all the anomalies survive. The Fano factor F_{RL} can be negative for certain ε and temperature high enough. Since the Fano factors for the three junctions are not equal, the charge conservation condition is not fulfilled. Both F_{L0} and F_{RL} become 1 for $\varepsilon \rightarrow -\infty$, 1/2 for $\varepsilon \rightarrow \infty$ and $F = 5/9$ for $\varepsilon = 0$. The striking difference between the Fano factors for the inner and outer junctions is that F_{0L} has no maxima or minima and just smoothly decreases from 1 to 1/2, whereas F_{RL} has two minima, one for $\varepsilon < 0$ and the other for $\varepsilon > 0$.

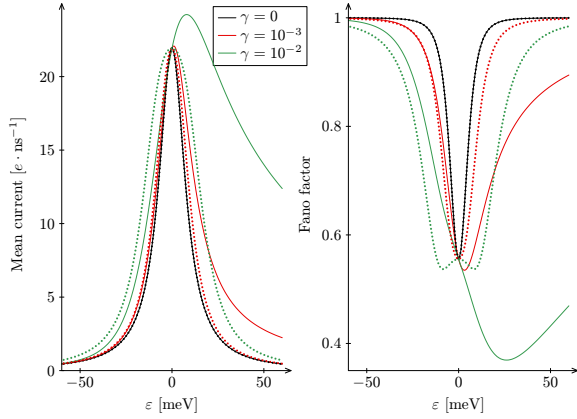


Fig. 4. Mean current and Fano factor vs. device bias ε for different values of damping coefficient γ . Solid lines correspond to the outer junctions without the RWA while dotted lines correspond to all junctions within RWA. Parameters: $\Gamma = 0.1/\hbar$ meV, $\Omega = 5$ meV, $\beta = 0.1$ meV $^{-1}$.

6.4. Rotating wave approximation

The rotating wave approximation (RWA) is understood as neglecting terms of the form $\sigma^\dagger \rho \sigma^\dagger$, $\sigma \rho \sigma$ and keeping terms of the form $\sigma^\dagger \rho \sigma$, $\sigma \rho \sigma^\dagger$ in the generalized master equation $d\rho/dt = \mathcal{L}\rho$ [2]. In our language, we mean $\sigma^\dagger \equiv |L\rangle\langle R|$, $\sigma \equiv |R\rangle\langle L|$ and, therefore, $|L\rangle\langle L| = \sigma^\dagger \sigma$, $|R\rangle\langle R| = \sigma \sigma^\dagger$. Application of the RWA to (7) leaves us with

$$\mathcal{L}_B^{\text{RWA}} = \begin{pmatrix} 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & 0 & 0 \\ 0 & 0 & 0 & -\gamma_p & 0 \\ 0 & 0 & 0 & 0 & -\gamma_p \end{pmatrix}.$$

The anomalous current (23) is now identically zero, since $[\mathcal{N}_L, \mathcal{L}_B^{\text{RWA}}] = [\mathcal{N}_R, \mathcal{L}_B^{\text{RWA}}] = 0$, and therefore the charge conservation is restored. The zero-frequency noise and the Fano factor for all junctions are equal. The mean current is

$$\langle I \rangle = \frac{e\Gamma\Omega^2\Lambda}{\frac{1}{2}\Gamma\varepsilon^2 + 3\Omega^2\Lambda + \frac{1}{2}\hbar^2\Gamma\Lambda^2}$$

and for the Fano factor we get

$$F = 1 - \frac{4\Omega^2 [\Lambda^2 (\Omega^2 + \frac{1}{2}\hbar^2\Gamma(\gamma_p + \Gamma)) + \frac{1}{2}\gamma_p\Gamma\varepsilon^2]}{[\frac{1}{2}\Gamma\varepsilon^2 + 3\Omega^2\Lambda + \frac{1}{2}\hbar^2\Gamma\Lambda^2]^2}.$$

These results are illustrated in Fig. 4.

We can see that negative Fano factor does not appear in this approach. Furthermore, all junctions give

identical results for both the mean current as well as the current noise. Thus, it seems that all problems are fixed. However, the physical content of our results has undergone great changes. We have given reasons why the mean current and the Fano factor curves should have emission-absorption asymmetry, but we obtain absolutely symmetric curves – the mean current gained the Lorentzian shape and the Fano factor has no suppression for $\varepsilon > 0$ due to the emission process. Because of these reasons we must reject the rotating wave approximation on the physical grounds.

6.5. Pauli master equation

Pauli master equation is yet another modification of the original GME which does not lead to formal inconsistencies. When the coupling to the leads Γ is small enough, which is the case in the regime we consider here, we can neglect the off-diagonal elements ρ_{12} , ρ_{21} of the density matrix in the system eigenbasis [10]. If we transform the Liouvillian of the original GME in the left-right basis (5) into the system eigenbasis $\{|0\rangle, |1\rangle, |2\rangle\}$, we can get the Liouvillian (or rate matrix) of the Pauli master equation immediately just by restricting ourselves to the subspace spanned by ρ_{00} , ρ_{11} , ρ_{22} which leads to

$$\mathcal{L}^{\text{Pauli}} = \begin{pmatrix} -\Gamma_{10} - \Gamma_{20} & \Gamma_{01} & \Gamma_{02} \\ \Gamma_{10} & -\Gamma_{01} - \gamma_\downarrow & \gamma_\uparrow \\ \Gamma_{20} & \gamma_\downarrow & -\Gamma_{02} - \gamma_\uparrow \end{pmatrix},$$

where

$$\begin{aligned} \Gamma_{10} = \Gamma_{02} &= \Gamma \frac{\Delta + \varepsilon}{2\Delta}, & \Gamma_{01} = \Gamma_{20} &= \Gamma \frac{\Delta - \varepsilon}{2\Delta}, \\ \gamma_\downarrow &= \frac{4\pi}{\hbar^2} \frac{\Omega^2}{\Delta^2} J(\Delta/\hbar) \frac{1}{1 - e^{-\beta\Delta}}, \\ \gamma_\uparrow &= \frac{4\pi}{\hbar^2} \frac{\Omega^2}{\Delta^2} J(\Delta/\hbar) \frac{1}{e^{\beta\Delta} - 1}. \end{aligned}$$

Identical results can be derived directly from a rate equation approach to the occupations of the eigenstates only with the rates determined by the Fermi golden rule. The current superoperators in the new Liouville subspace are given by

$$\mathcal{I}_{L0} = \begin{pmatrix} 0 & 0 & 0 \\ \Gamma_{10} & 0 & 0 \\ \Gamma_{20} & 0 & 0 \end{pmatrix}, \quad \mathcal{I}_{0R} = \begin{pmatrix} 0 & \Gamma_{01} & \Gamma_{02} \\ 0 & 0 & 0 \\ 0 & 0 & 0 \end{pmatrix},$$

while no analogy of the superoperator \mathcal{I}_{RL} exists on the chosen subspace.

For the mean current and the Fano factor between the dot and the lead, we get regardless of the choice of the left or right junction

$$\langle I \rangle = \frac{e\Gamma\Omega(\Omega\Lambda - \gamma_+\varepsilon)}{\frac{1}{2}\Gamma\varepsilon^2 + 3\Omega^2\Lambda - \Omega\varepsilon(2\gamma_+ + \gamma_-)}$$

and

$$F = 1 - 2\Omega \times \frac{2\Omega^3\Lambda^2 - \Omega^2\varepsilon(\gamma_p + \frac{3}{2}\Gamma)(3\gamma_+ + \gamma_-) - \gamma_+\varepsilon^3\Lambda}{[\frac{1}{2}\Gamma\varepsilon^2 + 3\Omega^2\Lambda - \Omega\varepsilon(2\gamma_+ + \gamma_-)]^2}.$$

These expressions differ from the result (27) and the corresponding expression for F_{L0}, F_{0R} for the full-space reduced density matrix in the second or the first order of Γ , respectively, i.e., they both agree in the lowest order in Γ as should be expected. Thus, for small values of Γ we obtain very good agreement between the two approaches (curves are almost indistinguishable from the solid lines in Fig. 2). This finding finally justifies the results obtained by the full approach as physically plausible although it does not yield any hints where the problem of the full approach might lie nor does it say anything reliable for larger Γ 's.

7. Discussion and conclusions

We have presented our results for the dissipative DQD system and explicitly pointed out the paradoxes stemming from the Markovian GME description of this system, such as charge conservation breaking between different junctions or unphysical negative values of the current noise. The weak coupling prescription is known to possess severe conceptual problems [2,17] including the ambiguity of the choice of the kernel (e.g., direct weak coupling prescription à la Bloch-Redfield vs. RWA/secular approximation on top of that), breaking of positivity of the reduced density matrix within the Bloch-Redfield formalism, and breaking of the equations of motion (Ehrenfest theorem) within the RWA [18]. Thus, in some sense one shouldn't be surprised to find similar inconsistencies in the DQD study. Yet, there are also important differences between the above mentioned effects and the present findings such as relative importance of the various discrepancies even for very small coupling constants, i.e., significantly increased sensitivity of these effects

with the presence of the other bath. This indicates that the phenomena encountered here may be going beyond the “standard” weak coupling paradoxes and are specific to multiple-bath setups.

This conjecture seems to be supported by further facts: the charge conservation is fulfilled without the presence of the dissipative bath which is a necessary condition for the occurrence of the breaking. RWA applied to our system expectedly breaks the equations of motion (physically incorrect symmetry between the emission and absorption processes), however, the full Bloch-Redfield kernel not only disobeys the positivity (negative noise) but also breaks the equations of motion (charge conservation breaking) which does not seem to follow the standard weak coupling behavior. The multiple-bath dissipative systems have been known as challenging for quite long but at the same time they are characterized by physically interesting and sometimes counterintuitive behavior [19,20] which can be also responsible for the present findings. Moreover, as a part of the diploma thesis of the first author [15] other systems were studied, e.g., the energy transport in two linearly coupled harmonic oscillators being an exactly solvable analog to the dissipative DQD but no similar phenomenon was observed there. This leads to speculations that these findings are determined not only by the simultaneous presence of more baths but also by the non-Gaussian character of the associated noise missing in the linear systems.

To sum up, although the reported issue resembles the notorious difficulties and paradoxes of the weak coupling theory, neither its exact origin nor possible cures have been uniquely identified so far. Yet, we present our findings to the community in order to draw attention to this open nontrivial problem which may be, apart from its relevance for the particular physical system under study, of interest in the broader context of open dissipative quantum systems. In particular, our findings raise the questions of the development of charge-conserving approximation schemes within the generalized master equation approaches analogous to their non-equilibrium Green's function counterparts and of general understanding of dynamics of quantum systems coupled to multiple baths.

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